

Docket No.: 294695US0PCT  
(PATENT)

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Patent Application of: Junichi Nakamura *et al.*

Application No.: 10/588,318

Confirmation No.: 5709

Filed: December 26, 2006

Art Unit: 1793

For: COATING, AQUEOUS COATING MATERIAL  
AND PROCESS FOR PRODUCING COATING  
USING THE SAME, AND COATED ARTICLE  
WITH COATING

Examiner: Eli Mekhlin

**DECLARATION UNDER 37 CFR §1.132**

MS Amendment  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Dear Sir:

We, Junichi Nakamura and Motomi Tanaka, declare and state that:

1. We are citizens of Japan, residing at c/o Toyohashi Production Center, Mitsubishi Rayon Co., Ltd., 1-2, Ushikawadori 4-chome, Toyohashi-shi, Aichi 440-8601, Japan. We are currently employed by MITSUBISHI RAYON, CO., LTD., and have been engaged in research on the production of coating and aqueous coating material.

2. It is our understanding that the invention set forth in the pending claims of the above-identified patent application (for which we are co-inventors) relates to:

(a) a coating comprising a coating component containing a polymer (I) and colloidal silica (II) having an average particle size of 60 nm or less, wherein colloidal silica (II) is contained with 0.5-20 parts by mass in solid content for 100 parts by mass of polymer (I) and an area of colloidal silica exposed at a coating surface occupies 35% or more of the coating surface;

(b) an aqueous coating material comprising a polymer (I) which comprises 0.1-20 parts by mass of a radical polymerizable monomer unit containing hydrolysable silyl group (a) and 80-99.9 parts by mass of another copolymerizable monomer unit (b) (total contents of (a) and (b) are 100 parts by mass), colloidal silica (II) having an average particle size of 60 nm or less, and at least one kind of an anionic surfactant shown by (III) consisting of sulfate salts of polyoxyalkylene aryl ether, sulfate salts of polyoxyalkylene alkylaryl ether, formalin condensates of sulfate salts of polyoxyalkylene aryl ether, and formalin condensates of sulfate salts of polyoxyalkylene alkylaryl ether and a nonionic surfactant, wherein colloidal silica (II) is contained with 0.5-20 parts by mass in solid content for 100 parts by mass of polymer (I);

(c) an aqueous coating material comprising polymer (I) which comprises 4-20 parts by mass of a radical polymerizable monomer unit containing hydrolysable silyl group (a) and 80-96 parts by mass of another copolymerizable monomer unit (b) (the total contents of (a) and (b) are 100 parts by mass) and colloidal silica (II) having an average particle size of 60nm or less, wherein colloidal silica (II) is contained with 0.5-20 parts by mass in solid content for 100 parts by mass of polymer (I);

(d) aqueous coating material comprising 0.5-20 parts by mass in solid content of colloidal silica (II) having an average particle size of 60 nm or less and 0.5-20 parts by mass of an organic solvent having less than 0.8 of a distribution coefficient of octanol/water;

(e) a process for producing a coating of the present invention comprises applying the aqueous coating material according to any one of (a) to (d), wherein a coating comprises a coating component containing polymer (I) and colloidal silica (II) having an average particle size of 60 nm or less, in which colloidal silica (II) is contained with 0.5-20 parts by mass in solid content for 100 parts by mass of polymer (I) and the area of colloidal silica exposed at the coating surface occupies 35% or more of the coating surface; and

(f) a coated article comprises the coating according to (a).

4. In order to demonstrate that the composition according to the present invention exhibits unexpected results, the following experiments were performed by us:

## EXPERIMENTS

### 1. PURPOSE

In order to show differences between the present invention and the invention of JP 09-165554 (YANAGASE et al.) cited by the Examiner in the first Office Action, an aqueous coating composition described in Example 5 of JP 09-165554 is replicated and occupied areas of colloidal silica exposed at a coating surface obtained by the aqueous coating composition are determined.

### 2. EXPERIMENTS AND RESULTS

#### (1) Production of Aqueous Coating Composition      Experiment Date: June 13, 2007

A graft block polymer emulsion was produced in accordance with the procedure of Example 5 of JP 09-165554.

The unavailable raw material was substituted with available raw materials, that is, SNOWTEX 30 (particle size: 10-20 nm, solid content: 30%) was substituted with SNOWTEX 20 (particle size: 10-20 nm, solid content: 20%). Though the mass ratio of deionized water for polymerization was changed in consideration of reaction scale, the characteristics of the graft block polymer emulsion themselves are not affected.

#### [Production of Silicone Emulsion]

The following raw material compositions were premixed with a homomixer, and the pre-mixture was forced-emulsified at 200 kg/cm<sup>2</sup> with a homogenizer to obtain a raw material pre-emulsion. Then, 55 parts by mass of water and 5 parts by mass of dodecylbenzene sulfonic acid were poured into a flask equipped with an agitator (stirrer), a reflux cooling pipe, a thermal control unit, and a titration pump. While keeping the inner temperature of the flask at 85°C under agitation, the raw material pre-emulsion was titrated for 4 hours. After the titration, a silicone emulsion was prepared by polymerizing for one hour, cooling, and adding ammonia with an equal molar content of dodecylbenzene sulfonic acid. The solid content thereof was 23.0% by mass.

## Raw material compositions

Mixture of trimer to heptamer of cyclic dimethylsiloxane oligomer	95 parts
$\gamma$ -Methacryloyloxy propyl trimethoxysilan	5 parts
Deionized water	250 parts
Sodium dodecylbenzene sulfonate	0.4 parts
Dodecylbenzene sulfonic acid	0.4 parts

## [Production of Graft Block Polymer Emulsion]

The following first raw material mixture was added into a flask equipped with an agitator, a reflux cooling pipe, a thermal control unit, and a titration pump. The inner temperature of the flask was raised to 750°C, and 0.48 parts by mass of potassium persulfate was added.

## First Raw Material Mixture

SiEm (solid content: 20 parts)	87 parts
Deionized water	47 parts
Sodium dodecylbenzene sulfonate	1 part

After 5 minutes of addition of potassium persulfate, the following second raw material mixture was titrated for 4 hours. The inner temperature of the flask was maintained at 70°C during titration, further maintained at 70°C for 1 hour after titration was completed, and then raised to 80°C and maintained for 1 hour.

## Second Raw Material Mixture

Methyl methacrylate	33.6 parts
Normal-butyl methacrylate	33.6 parts
2-Ethylhexyl acrylate	8.8 parts

Methacrylic acid	1.6 parts
Diacetone acryl amide	2.4 parts

Afterwards, the inner temperature of the flask was cooled down to room temperature, 1.12 parts by mass of 28% aqueous ammonia was added, and then the following raw materials were added in order to obtain an aqueous coating composition A.

SNOWTEX 20 (solid content: 12 parts)	60 parts
Adipic dihydrazide	1.52 parts
Butyl cellosolve	20 parts

## (2) Preparation of Coating (coating film)

The aqueous coating composition A provided by the above production method was coated on a PET film with a bar coater (#60) at 40°C and was dried at 130°C for 5 minutes. Furthermore, this aqueous coating composition A was dried at room temperature for 1 day. This coating was used as a coating plate for evaluation.

## (3) Measurement Method of Occupied Area of Colloidal Silica Exposed at Coating Surface

The occupied area of colloidal silica exposed at the coating surface obtained by the above process was determined by image processing of a secondary electron image of a scanning electron micrograph. The software used for the electron micrograph scan condition and image processing is shown as follows.

JSM-6340F type field emission scanning electron microscope manufactured by JEOL Ltd.

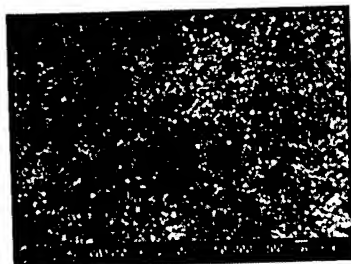
Acceleration voltage: 2.5kV

Magnification: 50,000 times / image processing analysis

Planetron Inc., Image-Pro Plus

Analysis target minimal area:  $1\text{e-}5\ \mu\text{m}^2$

The obtained electron micrograph is as follows.



#### (4) Evaluation Results

The aqueous coating material of Example 16 of the present invention and the aqueous coating composition A prepared by replicating Example 5 in Japanese Unexamined Patent Application No. 9-165554 (hereinafter, refers to JP 9-165554) were compared and evaluation results thereof are shown as follows.

		Present invention Example 16	Aqueous coating composition A JP 9-165554, Example 5
Polymer (I)		100 parts by mass	100 parts by mass
Colloidal silica	Particle size	10 - 20 nm SNOWTEX O	10 - 20 nm SNOWTEX 20
	Ratio	5 parts by mass	12 parts by mass
Anionic surfactant selected from (III) described in paragraph [0084] of present invention		CP12Na	Not used <sup>*1</sup>
Nonionic surfactant		EMULGEN 1150S-70	Not used
Occupied areas of colloidal silica exposed at coating surface (%)		97% (Attachment 1)	16% (Attachment 2)
Stain-resistance		◎	×

- \*1 Dodecylbenzene sulfonic acid was used. It is an anionic surfactant, but it does not have polyoxyalkylene, so it differs from anionic surfactants (III).

In the coating obtained from the aqueous coating material A which was prepared with reference to example 5 of JP 9-165554, occupied areas of the colloidal silica exposed at the coating surface are 16%. Furthermore, this coating formed a heteroaggregation structure.

On the other hand, the coating of example 16 of the present invention contains only 5 parts by mass of colloidal silica which was below a half amount of colloidal silica used in example 5 of JP 9-165554; however, occupied areas of the exposed colloidal silica on the coating surface are 97%.

For Declaration, add:

1. I am a citizen of Japan, residing at c/o Toyohashi Production Center, Mitsubishi Rayon Co., Ltd., 1-2, Ushikawadori 4-chome, Toyohashi-shi, Aichi 440-8601, Japan. I obtained a master's degree from Yokohama National University, where I studied Engineering. I am currently employed by MITSUBISHI RAYON, CO., LTD., and have been engaged in research on the production of coating and aqueous coating material since 2000.

2. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made may be punishable by fine or imprisonment, or both, and that such willful false statements may jeopardize the validity of this Application for Patent or any patent issuing thereon.

Dated: 2010. 2 . 23

Respectfully submitted,

By Junichi Nakamura  
Junichi Nakamura



For Declaration, add:

1. I am a citizen of Japan, residing at c/o Toyohashi Production Center, Mitsubishi Rayon Co., Ltd., 1-2, Ushikawadori 4-chome, Toyohashi-shi, Aichi 440-8601, Japan. I obtained a master's degree from Hiroshima University, where I studied Engineering. I am currently employed by MITSUBISHI RAYON, CO., LTD., and have been engaged in research on the production of coating and aqueous coating material since 1998.

2. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made may be punishable by fine or imprisonment, or both, and that such willful false statements may jeopardize the validity of this Application for Patent or any patent issuing thereon.

Dated: 2010. 2. 23

Respectfully submitted,

By Motomi Tanaka  
Motomi Tanaka